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IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF :

OLEG STENZEL, ET AL : GROUP: 1793

SERIAL NO: 10/542,763 :

FILED: JANUARY 17, 2006 : EXAMINER: PARVINI, P.

FOR: HIGHLY DISPERSIBLE SILICA :  
FOR USING IN RUBBER

DECLARATION UNDER 37 CFR 1.132

COMMISSIONER FOR PATENTS  
ALEXANDRIA, VIRGINIA 22313

SIR:

Now comes ANDREW WEHMEIER who deposes and says that:

- (1) I am an inventor of the above-identified invention.
- (2) Since 1998 I have been employed by EVONIK as a researcher engaged in the study of PRECIPITATED SILICAS AND RUBBER REINFORCEMENT.
- (3) I have considered the Office Action of March 4, 2008 and the Esch et al and Boyer et al patents cited therein.
- (4) The following information provides details of the preparation of the vulcanizable rubber composition of Example 3 of the Esch et al patent of record and a vulcanized rubber product prepared therefrom. Also provided are data involving the same tests for the rubber embodiment of the present invention identified as Example 1.5 on pages 33 *et seq* of the specification.

(5) Table 1 below provides the details of the mixing of ingredients for the preparation of a vulcanizable rubber formulation. The details of the mixture of ingredients are also provided by the "reference" formulation in Table 2.5 on page 40 of the specification. In the table, the abbreviation "phr" means parts by weight relative to 100 parts by weight of the raw rubber used.

Table 1

Substance	Phr	Article designation	Company
<b>1<sup>st</sup> step</b>			
Buna VSL 50/55-1	96.0	Basic mixing	Lanxess Europe GmbH & Co. KG; 51369 Leverkusen; Germany
Buna CB 24	30.0	S-SBR; oil-extended	Lanxess Europe GmbH & Co. KG; 51369 Leverkusen; Germany
Silica	80.0	dis-1,4-BR	
X 50-S	12.8	SI 69 (bis(3-(3,4-dethoxysilylpropyl)tetrasulfane) / carbon black of type N 330: 50% / 50%	Evonik Degussa GmbH; 45128 Essen; Germany
ZnO; RS RAL 844 C	3.0	ZnO	Amsperger Chemikalien GmbH; 50853 Cologne; Germany
EDENIOR ST1 GS	2.0	Palmitic-stearic acid; "Iodine number 1" stearin	Caltic Deutschland GmbH & Co. KG; 40231 Düsseldorf; Germany
Nafolen ZD	10.0	Aromatic plasticizer oil	Chemetall GmbH; 60487 Frankfurt a. Main; Germany
Vulkanox 4020 / LG	1.5	N-(1,3-Dimethylbutyl)-N'-phenyl-p-phenylenediamine (6PPD)	Rhein Chemie Rheinau GmbH; 68219 Mannheim Rheinau; Germany
Proteklor G 3108	1.0	Mixture of refined hydrocarbon waxes	Paramelt BV; 706875 Paramelt BV; NL 1704 RJ Heerhugowaard; The Netherlands
<b>2<sup>nd</sup> step</b>			
Step 1 batch		Lasting / remill step	
<b>3<sup>rd</sup> step</b>			
Step 2 batch		Finish mixing	
Vulkacit D	2.0	N,N'-Diphenylguanidine (DPG)	Rhein Chemie Rheinau GmbH; 68219 Mannheim Rheinau; Germany
Vulkacit CZEG-C	1.5	N-Cyclohexyl-2-benzothiazole sulfenamide (CBS)	Rhein Chemie Rheinau GmbH; 68219 Mannheim Rheinau; Germany
Perfakt TBZTD	0.2	Tetrabenzylthiuram disulfide (TBZTD)	Flexsys N.V./S.A., Woluwe-Garden; B-1932 St. Stevens-Woluwe; Belgium
Ground sulfur	1.5	Finely divided sulfur according to Ph Eur, BP	Merck KGaA; 64271 Darmstadt; Germany

The general method for manufacturing rubber mixtures and their vulcanized derivatives is described in the "Rubber Technology Handbook" by W. Hofmann, Hanser Verlag, 1994. The specific mixing conditions for the composition described in Table 1 above are specified in the following Table 2.

Table 2

<b>1<sup>st</sup> step</b>		Brabender 350 S mixer, filling level 0.73, 70 rpm, chamber temperature 70 °C, friction 1 : 1.11 plunger pressure 5 bar
0.0 to 0.5 minutes 0.5 to 1.5 minutes 1.5 minutes 1.5 to 2.5 minutes 2.5 minutes 2.5 to 3.5 minutes 3.5 minutes 3.5 to 5.0 minutes  5.0 minutes	Polymers 1/3 silica, X 50-S Clean 1/3 silica Clean 1/3 silica, ZnO, stearic acid, oil, Vulkanox 4020, Protektor Clean Mix; with speed variation if necessary, in order to reach the ejection temperature Discharge batch (batch temperature 145 °C to 155 °C) and distribute on roll: Cut in and fold over 3 ° on left, 3 ° on right, turn over 5 ° for narrow roll nip, 5 ° for broad roll nip Draw out a rolled sheet	
<b>24 hours intermediate storage at room temperature to step 2</b>		
<b>2<sup>nd</sup> step</b>		Brabender 350 S mixer, filling level 0.71, 80 rpm, chamber temperature 80 °C, friction 1 : 1.11 plunger pressure 5 bar
0.0 to 2.0 minutes 2.0 to 5.0 minutes 5.0 minutes	Plasticize batch from step 1 Maintain batch temperature at 150 °C by speed variation Discharge batch (batch temperature 145 °C to 155 °C) and distribute on roll: Cut in and fold over 3 ° on left, 3 ° on right, turn over 5 ° for narrow roll nip, 5 ° for broad roll nip Draw out a rolled sheet	
<b>4 hours intermediate storage at room temperature to step 3</b>		
<b>3<sup>rd</sup> step</b>		Brabender 350 S mixer, filling level 0.69, 50 rpm, chamber temperature 60 °C, friction 1 : 1.11 plunger pressure 5 bar
0.0 to 0.5 minutes 0.5 to 2.0 minutes 2.0 minutes	Batch from step 2 Constituents of the 3 <sup>rd</sup> step Discharge batch (batch temperature 90 °C to 110 °C) and distribute on roll: Cut in and fold over 3 ° on left, 3 ° on right, turn over 5 ° for narrow roll nip, 5 ° for broad roll nip Draw out a rolled sheet in the thickness necessary for preparation of the test specimens	
<b>12 hours intermediate storage at room temperature until vulcanization of the test specimens</b>		

Table 3 below identifies the standard test procedures employed in order to determine the values of the physical properties of the vulcanized rubber composition of Example 3 of the Esch et al patent.

Table 3

Physical testing	Standard / Conditions
<b>ML (1+4), 100 °C, 3<sup>rd</sup> step (MU)</b>	DIN 53523/3 ISO 667
<b>Vulcameter test, temperature, 165 °C, deflection 0.5 °</b>	DIN 53529/3 ISO 6502
<b>MDR rheometer</b>	
<b>M<sub>L</sub> (dNm)</b>	
<b>t 90% (minutes)</b>	
<b>t 80% - t 20% (minutes)</b>	
<b>Tensile test on bar S 1, 23 °C (median values from 3 bars)</b>	DIN 53504, ISO 37
<b>Modulus 200% (MPa)</b>	
<b>Modulus 200% / modulus 50% (-)</b>	
<b>Ball rebound 70 °C (%)</b>	DIN EN ISO 8307, drop height 500 mm, steel ball, d = 19 mm, 28 g
<b>DIN abrasion, 23 °C, 10 N force (mm<sup>3</sup>)</b>	DIN 53516
<b>Viscoelastic properties, 50 N preliminary force and 25 N amplitude force, 16 Hz Temperature equilibration time 5 minutes, recording of measured values after 30 s of test time tan δ, 60 °C (-)</b>	DIN 53513, ISO 2856

Table 4 below presents the data obtained for the measured property values identified in Table 3 above.

Table 4

		US 5846506 Example 3	OZ 6287 Example 1.5
ML (1+4), 100 °C, 3 <sup>rd</sup> step	MU	90	88
MDR: 165 °C; 0.5 *			
M <sub>L</sub>	dNm	3.5	2.8
t 90%	min	9.7	6.4
t 80% - t 20%	min	3.2	2.1
Vulcanization time (165 °C)	min	20	20
Modulus 200%	MPa	8.0	11.9
Modulus 200% / modulus 50%	--	5.0	7.4
DIN abrasion; 10 N	mm <sup>3</sup>	77	63
Ball rebound; 70 °C	%	65.8	69.3
Zwick: 16 Hz; 50 N ± 25 N			
tan δ, 60 °C	--	0.134	0.109

(6) It is clear from the data presented in Table 4 above that the compound rubber material of Example 1.5 of the present invention exhibits a profile of superior rubber values in comparison to the compound rubber material of Example 3 of the Esch et al patent which is based on a precipitated silica having a Sears number (V<sub>2</sub>)/ BET ratio (0.085 ml/(5m<sup>2</sup>) outside the range of 0.150 to 0.280 ml/(5m<sup>2</sup>) that is presently claimed for the silica of the invention. The data in the table above show a better Mooney viscosity M<sub>L</sub> value in the MDR test. This indicates a superior processing property of the compounded rubber material within the scope of the present invention. Desirably, the vulcanization time t 90 % and the vulcanization rate t 80 % - t 20 % are drastically reduced over the values of the compared composition of Esch et al. These greatly improved raw-mixture properties can be attributed, in particular, to the higher and better ratio of Sears number to BET surface area, since hereby faster and more effective hydrophobing, and consequently greater binding capacity of the silane to the silica is possible. The greatly improved properties of the vulcanized derivative

can also be attributed for the most part to this analytical characteristic. For example, the improved binding of the silica leads to better reinforcement of the vulcanized derivative in the tension test, as can be observed in the higher value of Modulus 200 % and the higher Modulus 200 %/50 % reinforcement factor. As a consequence, the DIN abrasion is improved by more than 18 %. The hysteresis behavior, which correlates directly with the rolling resistance of a tire that is finished with the present running-surface compound, can also be improved simultaneously by more than 18 % with the precipitated silica of the invention (see  $\tan \delta$ , 60° C). This improvement is also confirmed by the ball rebound value at 70° C (0.109).

(7) The undersigned petitioner declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

(8) Further deponent says not.

25.06.2008

(Date)

